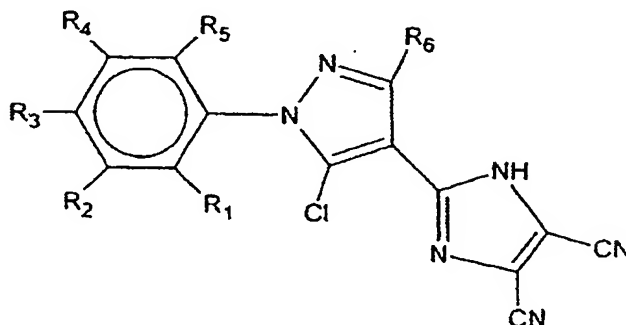


CLAIMS

1. A method for synthesizing 5-chloro-1-aryl-4-(4,5-dicyano-1H-imidazol-2-yl)-3-alkyl-1H-pyrazol
- 5 derivatives of general formula (I):



(I)

in which formula:

- R₁ to R₅, which may be identical or different, represent a group chosen from:
 - * a hydrogen atom,
 - * a halogen atom,
 - * a radical corresponding to the formula -(X)_n-R₇, in which X represents a group chosen from oxygen, sulfur, a sulfinyl radical and a sulfonyl radical, n is equal to 0 or to 1, and R₇ represents a linear or branched, saturated or unsaturated alkyl radical optionally substituted with one or more halogen atoms, which may be identical or different, this alkyl radical comprising 1 to 4 carbon atoms.
- R₆ represents a linear or branched, saturated or unsaturated alkyl radical comprising from 1 to 6 carbon atoms, optionally substituted with one or more halogen atoms, which may be identical or different, in which method a 1-aryl-3-alkyl-1H-pyrazolin-5-one derivative of formula (II) is used as starting product, this method being characterized in that:

- (a) in a first step, the pyrazolin-5-one derivative (II) is converted to the 1-aryl-3-alkyl-4-carboxaldehyde-5-chloropyrazol derivative of formula (IV) in one step by Vilsmeier treatment in the presence of POCl_3 and DMF,
- (b) in a second step, the aldehyde (IV) is converted to the 1-aryl-3-alkyl-4-[(2-amino-1,2-dicyanoethenyl)imino)methyl]-5-chloropyrazole corresponding to general formula (V) by condensation of the aldehyde (IV) with diaminomaleonitrile,
- (c) in a third step, the imine (V) gives the derivative according to general formula (I) via oxidative cyclization, which is carried out by treatment with a hypochlorite,
- according to the scheme represented in figure 2:

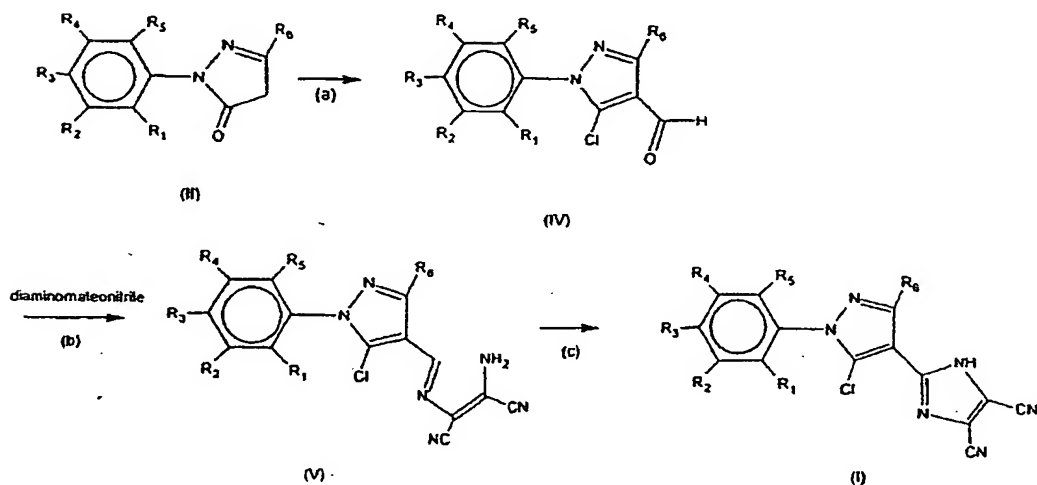


Figure 2

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2. The method as claimed in claim 1, characterized in that step (a) is carried out by treatment of the compound of formula (II) in DMF in the presence of 20 to 40 molar equivalents of POCl_3 , preferably 25 to 35 molar equivalents of POCl_3 , even more preferably 30 molar equivalents of POCl_3 .

25

3. The method as claimed in claim 2, characterized in that the (II)/DMF ratio is between 1 and 2, preferably between 1 and 1.5, even more preferably between 1 and 1.2.

5

4. The method as claimed in any one of claims 1 to 3, characterized in that step (b) is carried out in a solvent medium at a temperature of between 0 and 70°C.

10

5. The method as claimed in any one of claims 1 to 4, characterized in that step (b) is carried out in a methanolic medium with acid catalysis.

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6. The method as claimed in claim 5, characterized in that the catalyst is trifluoroacetic acid.

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7. The method as claimed in any one of claims 1 to 6, characterized in that step (c) is carried out by treatment of the compound corresponding to formula (V) with a hypochlorite chosen from an alkali metal or alkaline-earth metal hypochlorite or an alkyl hypochlorite, in a hydroxylated aliphatic solvent, at a temperature of between -5°C and 25°C, preferably of between 0°C and 5°C.

25

8. The method as claimed in claim 7, characterized in that sodium hypochlorite is used.

30

9. The method as claimed in any one of claims 1 to 8, characterized in that 1 to 5 molar equivalents of hypochlorite relative to the product (V), even more preferably 2 to 3 molar equivalents, are used.

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10. The method as claimed in any one of claims 1 to 9, characterized in that the product of general formula (V) is treated:

- in methanol,
- at a molar concentration of (V) ranging from 0.005 M to 0.1 M, advantageously from 0.01 M to

- 0.08 M, even more preferably from 0.02 M to 0.06 M,
- with a hypochlorite in quantity ranging from 1 to 5 molar equivalents, preferably from 2 to 3 molar equivalents, with respect to the product (V), this hypochlorite being in an aqueous solution having a concentration ranging from 1 to 5 M, preferably from 2 to 5 M.
- 10 11. The method as claimed in any one of claims 1 to 10, characterized in that steps (b) and (c) are carried out in a single step called (d), in the same reactor, without isolation of the intermediate product (V), in accordance with the reaction scheme which is
- 15 represented in figure 3:

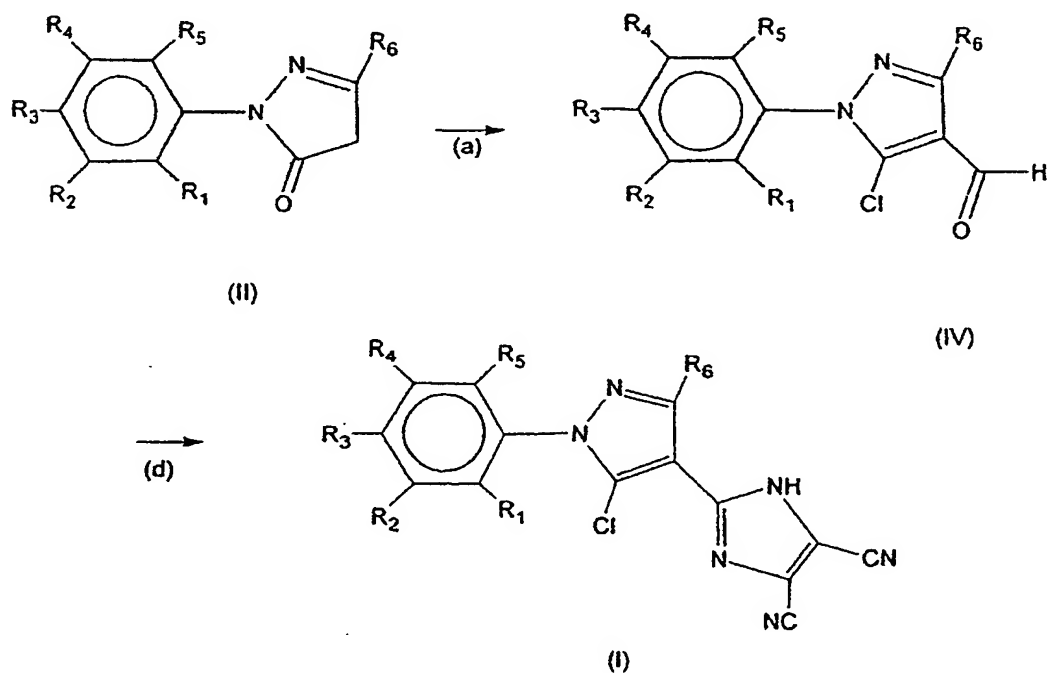


Figure 3

12. The method as claimed in claim 11, characterized in that
- 20 (a) in a first step, the pyrazolin-5-one derivative (II) is converted to the 1-aryl-3-alkyl-4-carboxaldehyde-5-chloropyrazol

derivative of formula (IV) in one step by Vilsmeier treatment in the presence of POCl_3 and DMF,

5 (d) in a second step, by successive treatment of the compound of formula (IV) with diaminomaleonitrile and then with a hypochlorite.

10 13. The method as claimed in claim 12, characterized in that step (d) is carried out in a hydroxylated aliphatic solvent medium, with, firstly, for the formation of the imine with diaminomaleonitrile, a molar concentration of substrate of between 0.15 and 0.2 M, with an acid catalysis, preferably provided by
15 trifluoroacetic acid, present in proportions of between 0.02 and 0.2 molar equivalent, and then, secondly, for the oxidative cyclization and the formation of the imidazolyl ring, dilution to a molar concentration of substrate of between 0.01 and 0.08 M, and the use of 2
20 to 3 molar equivalents of sodium hypochlorite having a concentration ranging from 2 M to 5 M.

14. The method as claimed in any one of claims 1 to 13, characterized in that, in formula (I), $n=0$.
25

15. The method as claimed in any one of claims 1 to 14, characterized in that one or more of the following conditions are met:

- 30 - R_1 to R_5 , which may be identical or different, represent a group chosen from:
- * a hydrogen atom,
 - * a halogen atom,
 - * a linear or branched, saturated or unsaturated alkyl radical R_7 , optionally substituted with
35 one or more halogen atoms, which may be identical or different, this alkyl radical comprising 1 to 4 carbon atoms,
- R_6 represents a linear or branched, saturated or unsaturated alkyl radical comprising from 1 to 4

carbon atoms.

16. The method as claimed in any one of claims 1 to 15, characterized in that one or more of the following conditions are met:

- R₁ to R₅, which may be identical or different, represent a group chosen from:

- * a hydrogen atom,
- * a chlorine atom,
- 10 * a linear or branched, saturated or unsaturated alkyl radical R₇, optionally substituted with one or more fluorine atoms, this alkyl radical comprising 1 to 4 carbon atoms,
- R₆ represents a radical chosen from methyl, ethyl, 15 tert-butyl and isopropyl.

17. The method as claimed in any one of claims 1 to 16, characterized in that the product of formula (I) is chosen from:

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5-chloro-1-(2,6-dichloro-4-trifluoromethylphenyl)-4-(4,5-dicyano-1H-imidazol-2-yl)-3-methyl-1H-pyrazol,

25 5-chloro-1-(2,6-dichloro-4-trifluoromethylphenyl)-4-(4,5-dicyano-1H-imidazol-2-yl)-3-isopropyl-1H-pyrazol,

5-chloro-1-(2,6-dichloro-4-trifluoromethylphenyl)-4-(4,5-dicyano-1H-imidazol-2-yl)-3-ethyl-1H-pyrazol,

30 5-chloro-1-(2,6-dichloro-4-trifluoromethylphenyl)-4-(4,5-dicyano-1H-imidazol-2-yl)-3-tert-butyl-1H-pyrazol.